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RESEARCH AND DEVELOPMENT TECHNICAL REPORT

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A REPORT ON THE DEVELOPMENT OF RARE EARTH-COBALT PERMANENT MAGNET TECHNOLOGY - JAPAN

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ELECTRONICS TECHNOLOGY & DEVICES LABORATORY



JULY 1981

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REVIEW OF PRESENT RARE EARTH PERMANENT MAGNET APPLICATIONS

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A REPORT ON THE DEVELOPMENT OF RARE EARTH-COBALT PERMANENT MAGNET TECHNOLOGY - JAPAN

INTRODUCTION

In connection with a request to chair a session and deliver a paper at the Fourth International Workshop on Rare Earth-Cobalt Permanent Magnets and Their Applications at Hakone, Japan, I took the opportunity to visit several laboratories and industrial manufacturers involved in the development of rare earth magnet technology and to assess the state of that technology. The following is a report on the Workshop and subsequent visits to various Japanese magnet manufacturers.

This was a very enlightening, fruitful trip. The Workshop at Hakone was well planned with many opportunities to make many useful contacts. After the Workshop, I was privileged to accompany Prof. Karl Strnat of the University of Dayton on a series of in-depth visits to several Japanese magnet manufacturing companies. Prof. Strnat d scovered the enhanced magnetic properties of the rare earth cobalt magnet materials in 1965 while employed at the Air Force Wright Patterson Materials Research Laboratories. Subsequently, his work at the University of Dayton under Air Force contracts was instrumental in initiating widespread development of these materials and their eventual use in many devices. Prof. Strnat is also the founder of the International Workshop series and was co-chairman of this Fourth International Workshop at Hakone. He is very much respected in Japan and honored as the father of this emerging new technology. Therefore, in his company, I was openly received by the various Japanese companies and was given access to information that ordinarily would have been closed to me. In what follows, I will attempt to put into perspective the state of the Japanese rare earth cobalt magnet industry as well as the development of new magnet material now taking place in Japan. However, before giving the highlights of the Workshop and the various company visits, certain general comments concerning the Japanese culture are necessary in order to understand the following discussion.

Succinctly, one can characterize the Japanese by saying that they care. They care about and respect people, nature and material things, and seek to shape or control them in a creative way. This care manifests itself in many ways: in the courtesy and helpfulness that the people show to one another and to strangers; in carefully sculpted trees and shrubs everywhere; in the lovely flowers and plant arrangements; in the careful maintenance of all buildings and mechanical equipment. Everything is kept spotless, many of the taxicabs have white seat covers, the drivers wear white gloves and are always shining their cabs. The trains and buildings are spotless with no graffitti or dirt to be seen. In the rush hours even though the trains are crowded the people are courteous and somehow display a great equanimity. This was also true in traffic jams. In nearly three weeks of a busy schedule in crowded cities, I saw only two overtly aggressive acts. In their very crowded cities it is clear that the Japanese have learned that courtesy, helpfulness, loyalty and team efforts have survival value. Graciousness, self respect, and mutual respect seemed to be present everywhere and may well form the basis for the much publicized, relatively stable management

employee relationship that seems to make Japanese companies so productive and creative. A great respect for the creative process permeates the society and is everywhere apparent whether it is in the tastefully dressed people, the lovely flower, plant and garden arrangements, the well-decorated store fronts or parks, the beautifully arranged food dishes or in many artfully created products. The people put in long hours, many working 10-hour days, six days a week. These traits were continually displayed in the various companies that we visited. These are the traits that make Japan, Incorporated so formidable!

Another important driver of Japanese productivity is the way Japanese companies are financed. Dr. Justin Bloom, Scientific Counsellor at the US Embassy, Tokyo, pointed out that whereas over 70% of United States business is stockholder financed, less than 30% of the Japanese corporations have such financing. In Japan most business ventures are financed through the banks with government sponsorship at the relatively low interest rates of 6-7%. Consequently, such businesses are not obligated to be responsive to stockholders, i.e., they are not under pressure to show a profit each year. They can afford to operate at a loss for a few years in a row while they develop a new technology. Furthermore, the lack of anti-trust legislation permits consortiums of big companies to pool their efforts to develop new technologies. An example is the cooperation of Toshiba, Mitsubishi, Hitachi and the Japan National Railway to develop a high speed. levitated train using either the technologies of superconducting or permanent magnets. Three different levitation schemes are presently being tested in prototype to develop a train capable of achieving velocities of 500 km/hr. Thus, Japanese companies are taking the technological risks while those in the United States are not. This statement is supported by the following item in the News from the Washington section of the IEEE Spectrum, June 1979, page 9:

U.S. SUPPORT FOR R&D FOUND LAGGING

"A grim picture of U.S. support of research and development was presented in Washington by spokesmen for an ad hoc organization that embraces 40 educational, scientific and technological societies. Harvard's president, Derek C. Bak, presided at a news conference on April 17 that cited National Science Foundation figures to show that from 1968 to 1978, R&D as a proportion of Gross National Product declined 20% in the U.S. while rising 15% in the Soviet Union, 16% in West Germany, and 20% in Japan. The ad hoc group also said that investment by industry in basic research, as a fraction of net sales, had gone down by 24% and that R&D, as a fraction of the Federal budget, had dropped by 40%. Along these same lines, Richard C. Atkinson, director of the National Science Foundation, told a Stanford University audience recently that "at a time when other nations have been increasing their emphasis on science and technology, we have been pulling back."

With this background in mind, I will report on the various items in my itinerary as listed in the Appendix.

TRAVELS:

21 May - Visit with the U.S. Army Science and Technology Center, Far East Office, Yokota Air Base

Personnel Contacted:

Colonel Reed, Commander US STC FEO
Major Baker, Staff Officer
Major A.H. O'Brien, Chief Communications-Electronic Branch
Mr. J.D. Busi, Technical Representative
Mr. R. Miller, Technical Representative

Major O'Brien picked me up at the Sanno early in the morning and drove me to the US STC FEO headquarters at Tokota, AB. After a welcome by Colonel Reed and a slide briefing on the mission of US STC FEO by Major Baker, I had some technical discussion with Jim Busi, Bob Miller and Major O'Brien. Mr. Busi specializes in electrochemistry and had had many contacts with people in our Power Sources Division, Electronics Technology and Devices Laboratory, when he had worked with the Foreign Science and Technology Center at Charlottesville, Va. He pointed out that the Japanese are now making big strides in all aspects of developing a hydrogen energy technology. The Matsushita Industrial Research and Technology Laboratory (MIRT) is a leader in this field and is actively developing small Stirling cycle heat pumps. Dr. Reji Sanno is studying various quaternary magnesium compounds having Ni, Zr and Cr substituents for hydrogen storage applications. The use of Cr gives dramatic control of the pressure plateaus as well as the rates of absorption and descrption. Dr. Suda of Kogaka University is a physical thermodynamicist active in the design of heat pump cycles. At the National Researchers Institute of Metals at Meguro, Tokyo, Dr. Y. Sasaki is also studying magnesium hydrogen storage alloys for use in heat pumps for automobiles. He has a materials research contract from the Daimler-Benz Company. Mr. Busi indicated that there will be an International Symposium on Metal Hydrides at Minakami Spa, Japan on 26-29 November 1979. Dr. Hirokiti Sasaki of Tohoku University at Sendai is chairman. Also, the 3rd World Hydrogen Energy Conference will be held in Tokyo, Japan, 27 June - 1 July 1980, and will be chaired by Prof. Tokio Ohta of Yokohama National University. I was later able to follow up on one of Mr. Busi's leads by visiting Dr. Y. Sasaki.

Mr. Bob Miller, who specializes in semiconductor technology and metallurgy, felt that the most important semiconductor work being done in Japan involved the development of the vertical junction transistor technology by Dr. Nishigawa at Tohoku University at Sendai. Such transistors were originally used for high power applications in high-fidelity audio equipment. However, Dr. Nishizawa has recently shown that these are also good high frequency devices. Devices now exist for operation at 3 GHz at 10 W with

100 W devices currently under development. The advantage of the vertical junction devices is that they exhibit no thermal runaway since their gain decreases with increasing temperature so that they are self-stabilizing. Mr. Miller indicated that he has written detailed reports on these developments. These reports are available through the Foreign Technology Office at Fort Monmouth. Miller also indicated that very large scale integrated (VLSI) technology is being pushed very hard in Japan with the five largest semiconductor companies cooperating under government sponsorship. A computercontrolled e-beam machine with quadrupole focusing and a resolution of better than one-half µm is already in operation. This machine was later mentioned during my visit to Toshiba. The present system is being pushed to the limits of its current computer capability and is being redesigned. Prototype 256 kilo bit (kb) memory chips have been fabricated, while 64 kb chips are in production. I asked him how the Japanese are handling the soft-mode failure problem. He said that they are well aware of the problem and are designing around it by the use of redundant circuitry.

Mr. Miller said that the Japanese effort for developing superconducting materials is similar to that in the United States. They are studying high transition temperature and high field materials for use in solenoids needed to develop the hydrogen fusion and levitated, high speed train technologies. Some work on Josephson junctions is being done at the universities but no big effort to produce a superconducting computer like IBM's is underway. In this regard the Japanese feel that they are five to ten years behind us as they have only been working with developing individual junctions and have not yet done very much with arrays of devices.

Major O'Brien has been following the levitated train technology developments in Japan. He told me about a prototype superconductively levitated train currently being tested which has achieved 200 km/hr on a 7-km test track. It is intended to extend the track to 15 or 20 km so as to attain the 500 km/hr velocities for which it has been designed. Levitated systems employing permanent magnets are also being developed in Japan using ferrite or rare earth cobalt materials. Major O'Brien told of attending an International Conference on Levitated Transport Systems held September 1978 in Japan. The meeting was dominated by the Japanese and Germans. Considerable political controversy seemed to exist between Japanese groups which favored different levitation schemes (attractive or repulsive) employing permanent magnets. Theoretical estimates presented indicated that such systems could be capable of achieving velocities of 900 km/hr before being limited by track tolerances.

Finally, it should be noted that Major O'Brien did an excellent job of helping to orient me to traveling around Tokyo and Japan. His help in making contacts with the various companies and his friendly advice contributed significantly to the success of my trip.

21-25 May - Fourth International Workshop on Rare Earth-Cobalt Permanent Magnets and Their Applications at Hakone

Personnel Contacted:

Professional Profession Control

Mr. J.G. Cannon, Molycorp, Inc., White Plains, NY

Dr. R.J. Churchill, Inland Motor Kollmorgen Corp., Radford, VA

Mr. A.W. Cornell, Hitachi Magnetic Corp., Edmore, MI

Mr. K.E. Davies, Rare Earth Products Ltd., Cheshire, England

Dr. P.H. Draper, Les Fabriques D'Assortiments Reuies, Switzerland

Mr. R.L. Fisher, Inland Motor Specialty Products Division, Radford, VA

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Dr. K. Kamino, Mitsubishi Steel Mfg. (o., Japan

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Mr. K. Kubo, Shin-Etsu Chemical Co., Itd., Japan

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Mr. T. Kurino, Society NonTraditional Tech. General Secretary of Workshop, Japan

Mr. Y-H Li, Applied Magnetics, Peoples Republic China

Dr. H. Maeda, Nat'l Research Institute for Metals, Japan

Mr. H. Millott, Data Products Corp., Woodland Hills, CA

Dr. A. Mohr, Robert Bosch GMBH, West Germany

Dr. K.S. Narasimhan, Crucible Inc. Res. Center, Pittsburgh, PA

Mr. H. Odajima, Sumitomo Special Metals Co., Japan

Mr. T. Ojima, TDK Electronics Co., Japan

Dr. M. Okada, Tohoku Univ., Japan

Mr. A.S. Rashidi, Hitachi Magnetics Corp., Edmore, MI

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Dr. T. Yoneyama, TDK Electronics Co., Japan

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Mr. T-D Soung, Rsch Inst. Iron & Steel, Peking Peoples Rep. China Prof. K.J. Strnat, Univ. of California, San Diego, La Jolla, CA

Mr. J. J. Blache, Rockwell International

Prof. M. Takahashi, Tohoku Univ., Japan

Dr. Y. Tawara, Shin-Etu Chemical Co., Japan

Prof. G. Thomas, Univ. of California, Berkeley, CA

Mr. G.L. Tilley, Union Oil Co. of Calif., Brea, CA

Dr. T. Tsushima, Electrotechnical Lab., Japan

Mr. M. Ueda, Sumitomo Metal Mining Co., Japan

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Mr. P. Wheeler, Wheeler Associates Inc., Elizabethtown, KY

Mr. L-Z Xu, Paotou Iron Steel Corp., Peoples Rep. China

Dr. E.J. Yadlowsky, Inland Motor Kollomorgen Corp., Radford, VA

Introduction

The Workshop was attended by nearly 200 people (of whom only about 10% were Americans). These were of very diverse backgrounds and included representatives of the rare earth refining industry, applications engineers, physicians and fundamental materials scientists. This mixture of talents leads to an interesting cross-fertilization that does not occur at the usual scientific meetings which tend to be more restricted in scope. Forty-six papers were presented at the twelve successive sessions which addressed the following topics (the number of sessions devoted to a given topic are in parentheses): Electrical applications (2); medical applications (2); magnetic bearings and other mechanical devices (1); magnetic properties (1); magnetic aftereffect (1); structures and coercivity (1); permanent magnets (3); and resources & refining of rare earth elements (1). Papers referenced in this section can be found in Proceedings of the Fourth International Workshop on Rare Earth-Cobalt Permanent Magnets, Hakone, Japan, May 22-24, 1979.

Applications

Prof. K.J. Strnat, University of Dayton, the discoverer of the rare earth permanent magnets (REPM), gave the opening plenary address in which he reviewed the history of the field and presented a summary of the latest device applications, material developments and future projections. This extensive overview nicely set the tone for the more detailed presentations that followed. He pointed out that while the initial uses around 1965 of SmCo₅ magnets were limited to replacing expensive platinum cobalt magnets (which then sold for \$3500 per kilogram) in military traveling tubes (TWT) and in specialized aerospace applications, the list of their present uses is rapidly growing. The outline of the present applications given by Prof. Strnat is quite impressive. It is shown in Table 1 with a few modifications to give the reader the present scope of REPM-based devices:

TABLE

REVIEW OF PRESENT RARE EARTH PERMANENT MAGNET APPLICATIONS

A. ELECTROMECHANICAL DEVICES

- 1. <u>Electric Motors</u>: Types DC (commutator and brushless), synchronous, induction start/sync. run; rotary and linear; continuous, torque, stepping. Geometries PM stator (conventional or ironless armature), PM rotor; radial or axial field (disc motor). Applications Watch and clock drives, aerospace gyros, momentum wheels, textile spinning turbines, servomotors, machine tool drives, (jet engine starter, electric vehicle propulsion, automotive accessory motors and starters under development).
- 2. Electric Generators: Types Brushless DC motor/generators, exciters, alternators, multi-phase sync. machines, pulse generators. Geometries PM rotor; radial or axial field; stator winding with or without iron. Applications Tachometers, satellite energy storage wheels, excitation of large sync. turbogenerators, aircraft turbine ignition, (400 Hz main aircraft generator/starter in development).

TABLE. REVIEW OF PRESENT RARE EARTH PERMANENT MAGNET APPLICATIONS (Cont'd)

- 3. <u>Electromechanical Actuators</u>: Linear Computer printers; magnetic head, laser mirror, recorder pen positioners. Rotary (Aircraft control surface (fin) actuators in development).
- 4. <u>Sound/Ultrasound Technology</u>: Microphones, earphones, loudspeakers, vibration senors, phonograph pickup.
- 5. <u>Measuring Instruments</u>: Moving-coil meters (core magnet), moving-magnet instruments.
- 6. <u>Electrical Switches</u>: Reed switches, snap-action relays, thermostats, eddy-current motor overspeed switch, Hall-effect switches (microswitch, automotive ignition).
- 7. Accelerometers/Gyroscopes: Military and commercial navigational/guidance systems.

B. MECHANICAL FORCE AND TORQUE DEVICES

- 1. <u>Couplings and Brakes</u>: Synchronous torque couplers, eddy current and hysteresis brakes, rotary-to-linear motion converter.
- 2. <u>Magnetic Bearings and Suspensions</u>: Passive Textile spinning turbines, ultra-centrifuges, watt-hour meters, record-player tonearm support, adjustable magnetic spring, (repulsive vehicle support). Partly active servoed system gyros, satellite momentum and energy wheels, laser beam scanner. (Turbo-molecular pumps, electromagnetic/electrodynamic vehicle levitation in development).
- 3. <u>Holding and Lifting</u>: Special purpose holding (elec. switching by flux displacement in development.) See also D. 1.2.
- 4. <u>Biomedical Devices</u>: Catheters, eyelid muscle assist, stoma seal, dental prostheses. (Implantable pumps and valves, head support harness in development).

C. MICROWAVE AND ION BEAM TECHNOLOGY

- 1. <u>Microwave Tubes</u>: TWT PPM focusing, klystrons, magnetrons, crossed-field amplifiers and backward-wave oscillators.
- 2. <u>Waveguide Devices</u>: Ferrite biasing in nonreciprocal waveguides, circulators etc., YIG resonance filters.
 - 3. Particle Accelerators: PM quadrupole lenses.
 - 4. Mass Spectrometer: Deflecting magnet in spacecraft instrument.

D. MISCELLANEOUS APPLICATIONS

- 1. Magnetic Locks: Key with magnets.
- 2. Magnetic Jewelry: Necklaces, clasps, earrings.
- 3. Electronic Chokes: Steady bias field.
- 4. Magnetic Bubble Memory: (Thin-film REPM for bias field under development).

Many of the device papers at the Workshop expanded upon the applications listed in Table 1 and demonstrated that REPM can achieve a significantly smaller device size for a given performance, simpler design with fewer parts, and advantages in machining and assembly which yield savings in production. In most designs the REPM have replaced electromagnets, Alnico magnets and in some cases even ferrites. As energy costs increase, designers of motors and of systems which traditionally use large electromagnets are now finding that operating expenses can now be significantly reduced by the increased efficiency offered by employing permanent-magnet fields. Rareearth magnets frequently offer the most energy-efficient design.

The most extensive development work in using the REPM has been done for motors and generators where the significant advantages that have resulted include: the possibility of novel machine geometries; improved cooling; smaller volume and weight; greater design flexibility and simplicity; and enhanced reliability. Several papers addressed the questions of new motor/generator designs. These were papers numbered 1, 2 and 3 in session I.

Microwave tubes built with REPM are outstanding examples of how the whole-system redesign approach can result in better, smaller and cheaper end products even though Alnico or ferrite magnets are replaced by the more expensive REPM materials. Three papers, numbers 2, 3, and 4 in session II, discussed such new backward wave tube, magnetron and TWT designs. Of particular interest was the fine paper (no. 2) by H.T. Soong and L.J. Kwang of the Southwest Research Institute of Applied Magnetics, Mienyang, Szechuan, Peoples Republic of China, on the design of O-type backward wave tubes for use at K band. This was the first indication that the Red Chinese have a well-developed rare earth magnet research and design effort in progress. Dr. Soong indicated that these tubes were now in mass production but refused to say for what application or where the tubes were being made. In this tube (Sm-Pr)Co₅ magnets having an energy product of 20 MGOe were being used in a well-designed step-type focusing system to produce a uniform axial field of 1400 Oe along a length of \sim 6.5 cm over a diameter of 2.8 cm. The ripple was less than 30 Oe along the axis. The magnetic circuit produced very low external leakage fields so that the tubes could be operated near magnetically sensitive instruments without any serious problems.

Papers II-3 and II-4 reported on the unique Japanese TWT and magnetron designs with REPM developed by the Toshiba and Hitachi companies. By incorporating the REPM inside the tube vacuum space, Toshiba has achieved a significant state-of-the-art improvement in their new 2M166 magnetron for microwave ovens. It has resulted in a tube having less than half the weight of the conventional tube used in a 600-W oven. The size of the tube was also reduced to nearly half the volume of the previous tube which employed ferrite. The weight of REPM used is only about 20 grams and represents less than 5% of the magnetron weight. The Hitachi people reported similar results.

A dramatic example of how the unique properties of high anisotropy, coercivity and energy products in the REPM permit radically new designs was given in paper II-1, "A New Generation of Samarium-Cobalt Quadrupole Magnets for Particle Beam Focusing Applications," by R.F. Holsinger of New England Nuclear and K. Halbach of Lawrence Berkeley Laboratory. A new class of permanent magnet quadrupole lenses for beam focusing in proton linear accelerators has been invented based upon the SmCo₅ material. An optimized

lens of simple design consists of 16 uniformly magnetized SmCo5 wedges of varying magnetization directions and contains no iron poles. Very high field gradients and pole-tip fields of about 10 kG have been achieved. The structure has only one-sixth the weight of a conventional electromagnetic lens of equivalent performance and of course consumes no power. The evolution of this work has been particularly satisfying to me, since Dr. K. Halbach credits me with sparking this new idea during a talk that I gave at the 3rd International Workshop on Rare Earth-Cobalt Permanent Magnets and Their Applications in 1978 at San Diego. In that talk and later conversations I had pointed out that more use should be made of a relatively obscure idea originated by W. Neugebauer who cleverly used the anisotropy of REPM materials for "guiding" flux around magnetic structures, thereby minimizing stray flux and enhancing the useful field. Subsequent interactions with Halbach, who is also a consultant to Los Alamos, led to his recommending to and our receiving money from Ballistic Missile Defense Advanced Technology Center (BMDTAC) in support of our Electronics Technology and Devices Laboratory (ET&DL) efforts to develop high coercivity and/or temperature compensated Sm₂Co₁₇-based magnets for further improvement of these quadrupole lenses.

Another group of devices made possible with the acvent of REPM are large magnetic bearings. These were discussed in session V, papers I through 6. Before the advent of rare earth magnets the only bearings of this type in production were for watt-hour meters and a lew clocks. These used magnets in repulsion for axial support of rotor weight. Since Earnshaw's theorem shows that complete three-dimensional stability cannot be attained in a bearing by means of permanent magnets alone, at least one degree of freedom always requires another bearing type such as a mechanical contact, an air cushion or an active electromagnetic system. The various papers presented detailed mathematical analyses of different bearing designs. Passive radial and axial PM bearings were described where repulsive or attractive forces (or both) are utilized. Mainly passive REPM bearings are now used in highspeed spinning turbines and ultra-centrifuges for isotope separation. Many servoed, partly or fully active electromagnetic bearing systems are now in development using Sm-Co magnets as crucial components. Most of these systems are for space applications. Other terrestrial applications being studied include large energy-storage flywheels and high-speed rotating electrical machinery, turbines, compressors, etc. Such bearings reduce friction, eliminate wear and the need for lubrication and cooling. However, Prof. Strnat pointed out that since these new bearings compete with the accepted technologies of oil and gas-pressure bearing and depend on unique concepts and materials, their widespread acceptance and use will probably be very

Sessions III and IV were devoted to various medical applications that employ REPM. These included various dental prosthetic devices, techniques for measuring blood flow electromagnetically, and various cytological studies of the effects of magnetic fields on the growth of normal and tumor cells. In paper III-2 no definite cytotoxic effects of SmCo5 or its magnetic field were noted, i.e., neither inhibition of cell growth or morphological changes of the FL strain of cells studied were noted. No chromosomal aberrations over the acceptable upper limit were noted. These findings support the adaptability and lack of cytotoxicity of SmCo5 magnets to the human body. They also showed a preference for using SmCo5 magnets plated with Cr for various implantation applications, e.g., for fixing dentures to gums. Initial inhibitory effects on the growth of several standard strains

of tumor cells were noted on exposure to a field of 6 kOe but the effects diminished with succeeding subcultivations. The mitotic activity was inhibited in the magnetic field and reached the minimum value 3-4 hours after removal of the cells from the field, and recovered almost completely after 24 hours. Further research on these effects was recommended.

In private conversations with Mr. T.E. Soung of the Peoples Republic of China I was told of a new magnetic acupuncture technique which employs rotating SmCo₅ magnets applied in the vicinity of the conventional acupuncture points. This method does away with the need for inserting needles at the various points. Mr. Soung gave me a copy of a publication in Chinese which describes work done during the period from 1973-1975. Success is claimed with such varied conditions as high blood pressure, bronchitis, asthma, skin problems, heart disease and vertigo.

Materials

The latter half of the conference, Sessions VI through XII, dealt with the various REPM materials, their fabrication and properties. Generally two types of magnets are distinguished by their production methods and physical characteristics. There are the sintered magnets and polymer-bonded (matrix) magnets. All sintered types (1-5 or 2-17; Sm, Ce, Pr or mischmetalbased) are hard and brittle and must be shaped by grinding or machining with diamond tools. They have fair electrical and thermal conductivities with good temperature/time stabilities for many applications. Polymer-bonded powder magnets made from the same alloys are also presently available. However, these offer less magnetic flux, poorer stability and lower maximum useful temperature (50-100°C) than their sintered counterparts. However, they can be pressed to size, easily machined or cut, are not brittle, and can even be made flexible or soft at the price of lower magnetic flux. Electrically and thermally they range from poor conductors to poor insulators. Their use is restricted to applications where ease of shaping and handling is more important than high energy, use is near room temperature and stability is not critical.

Certain topics or papers stand out as being of particular interest. These dealt with the questions of mechanical stability, the effect of oxygen content on coercivity, the effect of Ti, V and H_f substituents on the coercivity, and energy product of 2-17 compounds, new heat resistant bonded magnets, and new composite reinforced RE-Co permanent magnets. These will now be discussed briefly in turn.

During several private discussions the need was emphasized for good data on and approaches for improving the mechanical properties of the 1-5 and 2-17 type magnets so as to minimize losses due to fracture in handling or heat treatment. Very little work has been done on this topic in the West or in Japan. The only two papers which touched on this subject were both from the People's Republic of China. These were paper VI-4, "Thermal Expansion Anomalies of the Compounds RCo5" by Y.C. Yang, W.W. Ho and C. Lin of Peking (Beijing) University and paper XII-5, "The Anisotropic Thermal Expansion and the Fracture of Radially Oriented Toroid Specimens of the Rare-Earth-Cobalt Permanent Magnets" by T.D. Sun, J.H. Fhu and D.W. Wan of the Research Institute of Iron and Steel, Beijing.

In paper VI-4 the temperature dependence of the lattice constants of the compounds $CeCo_5$, $Ce(CoCu)_5$ and $Ce(CoCuFe)_5$ were reported for the temperature range 25° C to 600° C. $CeCo_5$ shows an Invar-type thermal expansion anomaly along the c-axis at the Curie temperature, T_c , i.e., a sharp increase in the slope of the C-axis lattice constant vs temperature curve occurs on heating above T_c . No such discontinuity is seen for the a-axis lattice constant which shows a linear temperature dependence over the whole range. This anomaly is considerably reduced on the partial substitution of Co atoms by Cu atoms. Neutron diffraction studies of the compounds $Y(CoCu)_5$ show that the Cu atoms preferentially substitute on the 2c-site. The reduction in the thermal expansion anomaly with Cu substitution is explained in terms of the Cu atoms acting to cut off very effectively the exchange interaction of the Co-Co atomic pairs along the c-axis.

Paper XII-5 dealt extensively with the results of a study of the fracture mechanism of radially oriented sintered SmCos toroids. It showed that the strong anisotropy of the thermal expansion controls the fracturing of the radial toroid after sintering. A detailed analysis in terms of the anisotropic values of the thermal expansion coefficients, moduli of elasticity, tensile strength and Poisson coefficients led to an expression for the critical degree of particle orientation with the c-axis along a radial direction. The parameter used to describe this orientation was $(\cos^2\theta)^{\frac{1}{2}}$, where θ denotes the angle between the c-axis of a crystallite and the radial direction along which it lies. Both the calculation and experiment show that the critical degree of the orientation, (CoS20)2crit, should not exceed 0.7. Experimentally, it was shown that the thermal expansion anisotropy could be decreased by employing a partial substitution of Cu for Co and a shift in composition toward the cobalt-rich (2-17 phase) region of the phase diagram. They found that the compositions Sm(Coo.80Cu_{0.14}Fe_{0.06})₇ and Sm(Co_{0.87}Cu_{0.13})₈ are almost completely isotropic and are ideal materials for making high performance, radially oriented toroids. The cause of the thermal expansion anisotropy in SmCo5 was fully discussed in terms exchange interactions between the various cobalt sites and their dependence on the various Co-Co lattice spacings. It was shown how the preferential substitution of Cu atoms on Co 2c-sites could explain the effective decrease in this anisotropy.

After Session VIII which dealt with the effects on coercivity of the various secondary phases in the 1-5 and Z-17 compounds, prolonged discussion took place concerning the effect of oxygen in these materials. The role of oxygen in these magnets is not very clear and has been controversial for some time. It seems to be present in the form of islands of samarium oxide SmyO3. C. Merget and M. Velicescu of the Goldschmidt Co., Essen, West Germany in Paper VIII-2 indicated that their magneto-optical Kerr effect and microprobe studies showed these inclusions occur as separate particles of 2-3µm size evenly distributed on the surface of the sample. The oxide particles are thought to be dead spaces that do not have any effect on the coercivity, though they lower the remanence and energy product. The oxide is not thought to be also present as the fine, submicron dispersion that would be required to produce the domain wall pinning needed to enhance the coercivity. But no one has been able to test this speculation definitively. Strnat felt that if the oxide were present as a fine dispersion of precipitates, it probably has a concentration less than the 4,000 to 5,000 ppm that M. Ishigaki and A. Higuchi of Sumitomo Special Metals Co, Japan have previously claimed yield on optimum intrinsic

coercivity in SmCo₅. However, in any case Strnat thought that it is indeed a finely dispersed precipitate of some kind, whether oxide of carbide that contributes to the high coercivity of $SmCo_5$ or Sm-mischmetal $Co_5-based$ magnets.

The question arose as to whether a very high purity SmCo5 magnet would still have a high coercivity. No one seemed to know. Strnat thought not. He doubted the possibility of achieving less than 200 ppm of oxygen on a commercial scale of production. Dr. K.V.S. Narasimhan of Colt Industries, Pittsburgh expressed the opinion that one would indeed be able to achieve high coercivity SmCo5 magnets with low oxygen content. He reported on efforts at Colt to produce low-oxygen content magnets under an Air Force Manufacturing Methods Technology contract. They have achieved magnets with energy products greater than 20 MGOe with oxygen concentration less than 2,000 ppm as compared to industrial standard magnets which usually have greater than 6,000 ppm of oxygen. The Colt results contradicted the Sumitomo results mentioned above. No one in the audience seemed to have any good ideas for resolving the discrepancy.

Recent problems with the international cobalt supply have limited the allocations to magnet manufacturers and have caused unprecedented increases in the price of cobalt. Since the rare earth magnets make much more effective use of the contained cobalt than do Alnicos, many of the Workshop talks on materials were concerned with the possibilities within the family of REPM of enhancing the magnetic energy per unit weight of cobalt above that obtainable even with SmCo5. Recent Japanese and Swiss work has indicated that energy product, (BH)_{max}, in excess of 30 MGOe can be achieved. In the Sm₂Co₁₇ and other 2-17 alloys, it is possible to substitute substantial amounts of Fe, Cu and also some Mn or Cr for a part of the Co and still be able to maintain or improve the performance of the material. Therefore, these factors have stimulated renewed materials development (of the Sm₂Co₁₇-based alloys), particularly by the Japanese.

Several papers were presented which dealt with the effects of various substituents such as Ti, Zr or Hf on the coercivity and energy products of REPM. Papers IX-3, and XII-3 by Inomata's group at the Toshiba Research and Development Center of Kawasaki, in collaboration with Gato, Sakurai and Ito of Tohoku University, dealt with "Magnetic Characteristics of Ce(Co,Cu,Fe,Ti)6.4 Magnets." Paper XII-4 by Yoneyama, Fukuno and Ojima of TDK Electronics Co., Ichikawa, Chiba was entitled "Magnetic Properties and Structures of Sm2(Co,Cu,Fe,Zr)17 Type Magnets," while paper XII-6 by Nezu, Tokunaga and Igarashi of Hitachi Metals Ltd., at Kumagaya, Saitama was "Sm2(Co,Fe,Cu)₁₇ Permanent Magnet Alloys with the Additive Element Hf." In all of these alloys the IV-A substituent acts to enchance the coercivity and thus the energy product. The exact mechanism for doing so is obscure. It was speculated that the presence of carbon in all of these compounds, as reported by Herget and Velicescu in paper VIII-2, points to the existence of a fine dispersion of the various IV-A carbines in the respective alloys. Such a dispersion might act as the source of the domain wall pinning defects needed to enhance the coercivity. In various discussions, it was generally agreed that it would be important for future work to determine whether the Ti, V, or Hf substituents go into solution and thus act on the crystal fields or whether they form the fine dispersion of oxides or carbines, referred to above, and act as pinning

Two important papers on resin-bonded magnets were presented. Paper XI-1 by Suzuki, Yamane and Kamino of the Mitsubishi Steel Mfg.Co., Tokyo and Hasegawa, Hamano and Yajima of Tohoku University, Oarai, Ibaraki was entitled "A Heat-Resistant R-Co Bonded Magnet." It dealt with a new type of heat resistant polymer-bonded R-Co magnet manufactured by using SmCo5 powder and a newly developed borosiloxane-based polymer that featured excellent thermal stability in air up to 400°C. Measurements of magnetic properties of the bonded magnet yielded $B_r = 6.3 \text{ kB}$, $iH_c = 6.4 \text{ kOe}$ and (BH) max = 9.3 MGOe. Also, the irreversible loss in spontaneous magnetization at -B/H = 3 was 2.5% for a 150° C heat treated sample and 5.5% for a sample treated at 200°C. Such characteristics are comparable to that of sintered SmCo₅ magnets. These excellent magnetic properties were confirmed to be due to (1) the heat resistance of the polymer, (2) the high content of the polymer in the magnet, over 2 wt%, (3) the high density, over 6.5 g/cc, of the magnet, and (4) the high coercivity of the SmCo₅ powder. The long term stability of this material has not been determined. These magnets are not yet commercially available.

Paper XI-2 by Shimoda, Kasai, and Teraishi of the Suwa Seikosha Co., Suwa, Nagano described a "New Resin-Bonded Sm-Co Magnet Having High Energy Product," in which Sm₂Co₁₇-based magnet material was used with an epoxy resin. High coercive force was obtained using a composition of $Sm(Co_{0.672}Cu_{0.08}Fe_{0.22}Zr_{0.028})$ where Z varied from 8.35 to 9.0. An _iH_c over 20 k0e was found for z = 8.35 with appropriate heat treatments. Initial magnetization of these magnets showed that the magnetization was controlled by domain wall-pinning. Since the ${}_{1}\mathrm{H}_{\mathrm{C}}$ did not depend on the particle size used, high density magnets ($\rho = 7.1 \text{ g/cm}^3$) were made by combining large particles with small ones to reduce the void content. The magnetic properties were $B_r = 8.5 \text{ kG}$, $_bH_c = 6.8 \text{ kOe}$ and $(BH)_{max} = 17.3 \text{ MGOe}$. These were exceptional results for a bonded magnet. Two very sophisticated production processes were described. In one method called "the whole die press forming method," the powders (magnet and resin) are packed, pressed and pulled out from the die in a single process. The resin is then cured to obtain the final magnet product. This method makes it possible to manufacture the magnets with excellent dimensional accuracy and requires only half the cost of the conventional process.

Another paper that aroused considerable interest was entitled "Composite Reinforced RE-Co Permanent Magnets," Paper XI-3 by P.H. Draper of Les Fabriques d'Assortiments Reunies, Le Locle, Switzerland. Draper described a new class of precipitation-hardened RE-Co permanent magnet alloys with improved mechanical properties which minimize the problems associated with the brittleness of currently available RE-Co materials. The microstructure of this class of materials consists of a small volume fraction of ductile cobalt dendrites, within a cobalt-copper-rare earth matrix and is produced by directions solidification. A pilot plant operation is presently being developed. Metallurgically the current alloys being commercially developed consist of three phases:

- the Co dendrite phase, occupying 10 volume %, and incorporating a large fraction of the other transition metal-elements.
- the 2-17 type phase, saturated with Sm and Cu and occupying about 70% of the volume.

The material is not fully in equilibrium at low temperatures according to the ternary phase diagram. However, the Co phase, once formed shows no measurable tendency to redissolve. The fracture energy of this material is five times greater than that of sintered SmCo₅ magnets. Consequently, the material exhibits greater machinability and can be turned and/or drilled using conventional hard metal techniques. Better surface quality and more precise dimensions can be obtained, particularly for small work pieces. The material is also less susceptible to thermal shock. The magnetic properties are modest compared to SmCo₅. Those presently attainable are: Br-8.0 kG, $_{\rm iH_C}$ = 5.5 kOe and (BH) $_{\rm max}$ = 14.0 MGOe. The major limitation results from the fabrication method which controls the form of the raw magnet material: a cylindrical bar, some tens of cms in length and about one cm in diameter with a unique magnetization axis along the length of the bar. Other cast forms are also possible using the techniques of directed solidification, but considerable development work will be required.

A special session was held on the Fundamental Problems of Rare Earth Cobalt Magnet Materials. It was chaired by Prof. Wallace of the University of Pittsburgh. He opened the session with a lengthy commentary of his own thoughts on the matter that I feel are worth preserving in this report, along with some of the replies that his thoughts prompted. Wallace felt that "the fundamental scientific base on which this technology is resting is very, very narrow indeed." Paraphrasing Winston Churchill, he said, "Never has so large a technology owed so much to such a small amount of basic science," and pointed out that the whole technology is now based on the characteristics of just the three materials: (Mischmetal)Co5,SmCo5 and Sm₂Co₁₇. He reviewed the properties required of a good permanent magnet material, namely: "a good remanence, a sufficiently high Curie temperature, and the more subtle feature of a strong magnetic anisotropy. alignment of the magnetic moments is maintained by the anisotropy energy, e.g., the crystal field interaction that operates between the several Co sublattices and the rare earth or Sm sublattice. The Co sublattice anisotropy in $SmCo_5$ is not very strong. It is not significant in producing good permanent magnet behavior. One needs the rare earth anisotropy, which makes the major contribution to the magnet anisotropy. What is wrong with the cobalt anistropy? Why doesn't it do the job? The anisotropy of Co in YCo5 and LaCo5 is quite high and is presumably due to the Co alone, but you can't take advantage of it there to attain good magnets. The anisotropy field of Sm in SmCo5, on the other hand, is quite high and you can take advantage of it. It is quite clear that the issue of the anisotropy that is developed out of the Co sublattices is very poorly understood." Wallace felt that the most revealing treatment of the problem has been done by Dr. Inomata at the Toshiba Research Center, "bu: he keeps his conclusions and his calculations locked up in a laboratory report written in Japanese because he is not confident that this really should be exposed yet to the full public. However, the issue of why there is the strong cobalt anisotropy sometimes and why it differs from the single ion anisotropy exhibited by the rare earth sublattice is not at all well understood.

Prof. Wallace emphasized the need for detailed band structure calculations before we will have adequate fundamental understanding of the electronic, magnetic and mechanical properties of the SmCo₅- and Sm₂Co₁₇-based materials. We don't understand the role of d-band sublattices in these materials.

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Wallace also pointed out that all the compounds are chemically dirty as well as being brittle. In fact, it is known that other substances become ductile when adequately cleaned up. He speculated that a very clean $SmCo_5$ material might be ductile and more amenable to simple manufacturing processes. Since these materials have a strong affinity for oxygen and carbon, the adverse mechanical properties may arise as a result of being so dirty. Wallace wondered about the properties that really clean, i.e., semiconductor-grade $SmCo_5$ and Sm_2Co_{17} compounds, might have.

Prof. Strnat replied that Joe Becker of GE, years ago showed that small single crystals of YCO₅, prepared carefully, attained a high coercivity and a (BH)_{max} of 28 MGOe, so that the Co anisotropy is not absolutely worthless. But it is somewhat more difficult to put to practical use than the rare earth crystalline anisotropy. Dr. Rashidi of Hitachi Magnetics said that "It would be nice to come up with a clean alloy, but it does not make a good magnet. All you would have would be a clean alloy." He indicated Hitachi has determined that in order to maintain repeatability of magnet properties a certain amount of oxygen is an absolute necessity. Lack of oxygen gives a shiny alloy, but the alloy does not make good magnets.

Strnat agreed with Rashidi and further said that higher coercivities might be attained in pure alloys from a scatter-disorder mechanism similar to that observed by Oesterreicher. But those coercivities seem to exist only at low temperatures but not at room temperatures. The high coercivity in any of the practical magnets seems to rely on some form of dirt, e.g., another intermetallic compound precipitated out, some type of stacking fault structure, and/or oxide or carbide precipitates. Possibly the zirconium or titanium carbides, that Dr. Valicesscu found, are important agents in causing the magnetic hardening. In the end you may need the dirt for the practical magnets.

Wallace, changing the subject, asked "Wouldn't it be nice not to be geared to use the Co? Wouldn't it be nice to have a SmFe5 alloy for example, that would behave substantially like SmCo5? A very interesting fundamental question is why don't the REFe5 compounds form." Of course ThFe5 does form, suggesting the possibility that there are some electronic factors involved. Wallace and one of his associates are now doing a band structure calculation for SmFe5 to see if by examination of the electronic band structure they might be able to ascertain what parameters might be manipulated to bring this material into existence.

Prof. Wallace also felt that there was a lack of a concerted effort on the part of the rare earth magnet community to explore other options. He pointed out that "Jeitsko, formerly of DuPont and now at the University of Giessen, recently reported on a number of rare earth iron phosphors which are 2-19 compounds and are materials which are potentially uniaxial. Their magnetic properties have not been investigated." He also noted that "the initial work on the structure of the 2-17 compounds was done many years ago in the Soviet Union by Gladisheskii who has continued to synthesize many potentially interesting new magnetic compounds. Their magnetic properties have also not been studied, even though they might be of interest for application. After all, the 1-5 compounds date back to the work of Novotony in Austria forty or fifty years ago. The 2-17's are also very old compounds. There are lots of things like these 2-19's or the compounds that Gladisheskii is providing or a number of other compounds that warrant investigation as

potential permanent magnet materials." It seemed to Wallace that the basic science that needed doing, collateral to the present technological effort, should be a strong follow-up on these new compounds. When Wallace asked for further suggestions for new materials from the audience, there were none.

Concerns About the U.S. Position

On the evening of 23 May, I attended a small party given by Joe Cannon, the Sales Manager of the Molybdenum Corporation of America, recently acquired by the Union Oil Company. In attendance were Prof. Ed. Wallace, Univ. of Pittsburgh; Dr. Russel Churchill, Vice President for Research, Inland Motors; John Blache, Autonetics Group, Rockwell International; Harold Millott, Dataproducts Corporation; and Jun Ikeda, Mitsubishi Corporation, the Union Oil/Molycorp representative in Japan. Joe Cannon expressed his concern about the relatively weak capability that the RE-Co magnet industry had in the USA, and wondered whether anything could be done about strengthening our All present were equally concerned. Churchill pointed out that Inland Motors had extensive plans for many new motor designs based on the REPM's, but that they were very reluctant to go into high volume-production because there were no American sources of the magnets with the capacity to supply their needs. Inland was very reluctant to make the large investments needed and then be primarily dependent upon foreign suppliers. Millot expressed similar concerns. The new high speed data printers that Dataproducts is making now depend upon Hitachi for the magnets. Dataproducts is also interested in developing US suppliers with large volume capability. The conversation explored the various reasons for the current situation, from the way businesses are structured in the USA, through the lack of venture capital during the past decade, the fact that many of the magnetic products are now being made almost exclusively in Japan so that a broadbased OEM market doesn't exist in the USA for magnets, to the severe shortage of trained magnetics people in the USA. No one seemed to have a good plan as to how the RE Co magnet effort could be revitalized in the States.

Summary

All in all this was a very productive, stimulating Workshop. Prof. Kaneko, Mr. Kurino and their colleagues are to be congratulated for the excellent arrangements and the gracious running of the Workshop. The many participants with whom I discussed the matter were well-pleased and looked forward to future Workshops. At this time tentative preparations are being made for a 5th Workshop in the USA and a 6th in Europe, while Prof. Ho Wen-Wang of Peking University has received permission from his government to hold the 7th meeting in China during 1982.

28 May - Visit to TDK Electronics Co., Tokyo

Personnel Contacted:

Mr. Y. Mano, General Manager, R&D Laboratory

Mr. M. Ishikawa, Product Manager, Navita Factory

Dr. T. Hiraga, Managing Director, TDK

Dr. T. Ojima, Senior Scientist

Dr. T. Hori, Chief Engineer, Magnet Div., Narita Factory

Dr. T. Honeyama, Research Scientist

After graciously being greeted by the above, Profs Strnat, Ray and I were ushered into a conference room where Strnat gave a one hour lecture summarizing the present state-of-the-art for RE-Co magnets. We then had discussions of technical problems of mutual interest. They were quite interested in trying to understand where the zirconium was going in their 30 MGOe, REC-30 alloy that is currently in production. Even though they have achieved a significant enhancement of the energy product they do not understand exactly why it is happening. They asked us our opinion of the role the zirconium was playing and where it was going in the crystal structure. Prof. Al Ray thought that the zirconium was probably occupying cobalt sites. His long experience with metallurgical processing led him to believe that the zirconium atom and all the other transition metal atoms, such as Ti, Nb, Ta, Hf that have been tried, will not go into a site where they rattle around. They tend to go into sites where they have to squeeze themselves in or can just barely make it. Therefore, he feels that it is into the Co-sites in the 1-5 compounds that these substituents go. In the 2-17 compounds they would go into only the "dumbbell" Co-sites. He feels that the rare earth sites are too large for the transition metal atoms to enter. However, he is not absolutely certainthat this is, in fact, occurring. Such atoms might also be going into a second phase. In that connection, I raised the question of Takahashi's work. During the Hakone meeting, I had met Prof. M. Takahashi of the Applied Physics Department of the Engineering Faculty, Tohoku University at Sendai. He privately described his very interesting work on dilute solutions of various transition elements and rare earths in pure cobalt. He had found some dramatic effects due to such substituents. They led to remarkable enhancements in the anisotropy energies when the dilute cobalt alloys were subjected to magnetic annealing. Apparently the effects are caused by a very finely dispersed second phase in pure cobalt. I wondered whether such a cobalt-rich dispersion might be present and act as the pinning centers in some of the alloys we were considering. Al Ray felt strongly that I was correct in assuming that there must be a finely dispersed phase present in these materials. He based his opinion on observations of large supercooling during differential thermal analysis experiments on the melts of 2-17 and 1-5 materials. The supercooling produces a supersaturated cobalt solution. The subsequent spontaneous heating, Ray feels, is indicative of a massive nucleation of a very finely dispersed second phase similar to that which I had intuitively suggested. So we asked the Japanese to look at the effects of a magnetic anneal on these materials to see if there is any enhancement of the anisotropy or coercivity that might indicate the presence of this finely dispersed cobalt precipitate. They indicated they would try this experiment. Ray also suggested that they put a thermocouple in their melts to see if they observe the DTA curve charactertistic of the massive supercooling effect.

The TDK people then took us on a tour of the magnetic research facilities. The laboratory was very impressive. Everything was clean and neat. They have a central data acquisition/computer facility for the online processing of experiments from all parts of the laboratory. Each room in the laboratory has a data acquisition/processing terminal which is connected to the computer. They had a complete materials characterization facility: x-ray diffraction, scanning and transmission electron microscopes, ESCA, and microprobe analysis. The completeness of the characterization facilities puts most of the United States government and industrial facilities to shame. They had a very fine Edwards ion-milling apparatus for

thinning down their specimens for transmission studies. I was very impressed by the fact that this relatively small industrial company has had the vision to invest so heavily in and fully staff such an analytical, characterization, magnetics measurements and data acquisition/processing operation to provide rapid feed-back to their materials development people.

Their magnetics laboratory was equipped with a large 20 kOe electromagnet used in obtaining magnetization and anisotropy field measurements. They have to extrapolate to high magnetic fields to estimate their anisotropy fields and intrinsic coercivity values. They use a rather simple arrangement to obtain high temperature magnetization data with their TEOL vibrating sample magnetometer. A quartz tube with a bifilar nichrome winding that is insulated with asbestos sheeting permits measurements to 1,000 °C. No water cooling is used.

Dr. Ojima gave me some samples of their 30 MGOe, REC-30 material so that we could do anisotropy and coercivity measurements in the range from 4.2 to 300K. They are not equipped to do such low temperature measurements.

We were told about the new TDK plant at Narita which opened in September 1978, and now employs about 100 people. It has 9,000 m² for magnet production and 7,500 m² for product assembly, e.g., RE-G necklaces, home drinking water deionizers and vaporizers. The RE-Co magnet production capacity is 10 tons/month. Presently, production is 3 tons/month, (40%, 1-5 material; 60% 2-17 material). The magnet types are: (1) SmCo5-made from (a) the Goldschmidt reduction-diffusion processed powder called TEOMAG and (b) the TDK cast alloy Sm 60/Co 40, (2) Mischmetal Co5, and (3) the REC-24 (parallel pressed) and REC-30 (transversely pressed) Sm2Co17-based magnets. According to Prof. Strnat, who subsequently visited the Narita plant, it was an all new, highly automated facility with the latest processing and machining equipment.

29 May - Visit to the Electrochemical Laboratory, Tanashi Branch, Tokyo

Personnel Contacted:

Dr. Tachiro Tsushima, Chief, Magnetic Materials Section

Dr. Tsugio Shibata, Senior Researcher

Dr. Takashi Horigome, Chief, Energy Systems Section

Dr. S. Ihava, Senior Researcher

The Electrotechnical Laboratory (ETL) is the largest national research organization specializing in electricity and electronics in Japan. Since its establishment in 1891, ETL has greatly contributed to the progress of science and technology by serving as a nucleus for R&D activities in Japan. ETL became affiliated with the Agency of Industrial Science and Technology in 1948. Since then many reorganizations and expansions have occurred in order to cope with the needs for rapid technological innovation. During fiscal year 1978 ETL had a staff of 768 of whom 573 were research personnel. The FY-78 budget was about \$32,000,000. At present ETL is primarily involved with the following four R&D areas: solid state physics and materials, information processing, energy, as well as standards and

measurements. ETL is also taking technical leadership in performing three national R&D programs: One is for MHD generation (1966-1982), another is for pattern information processing (1971-1980), and the last is the so-called "Sunshine Project" (1974-2000), in which ETL is engaged in R&D of solar and hydrogen energy technologies. ETL is also establishing fundamental technologies to serve as the basis for future national programs on space and ocean engineering.

We were hosted by Dr. T. Tsushima, Chief of the Magnetic Materials Section of the Electronics Technology Section of the Electronics Technology Division. At the Hakone Workshop, Dr. Tsushima had demonstrated his talents as a folksinger by entertaining us after the banquet dinner with ancient Japanese love songs. He is also quite a linguist. He speaks a flawless English and German. The Section had a permanent staff of eleven scientists, eight of whom were Ph.D.'s, and one secretary. It had no technicians, but had a temporary staff of eight consisting of one person from industry, three graduate students doing their PhD thesis at ETL and four undergraduate students. Dr. Tsushima indicated that it was quite common for ETL to supplement its staff with people from industry or the universities. The fields being studied by the Section were amorphous magnetic metals, rare earth cobalt magnets, amorphous thin films, magnetic semiconductors, thin film bubbles, magneto-optic studies, the liquid phase epitaxy and dynamic behavior of garnets. This is quite a range of materials. They had an impressive amount of equipment, all of it seemed to be functional. Some of it was dated but not too much so. Their laboratories were not as well equipped for analytical and characterization studies as the TDK laboratory. They have a unique high-temperature arc-imaging single crystal growth furnace. A high intensity halogen lamp is at one focus of a gold-plated elliptical cavity, while the single crystal is grown at the other focus by slowly pulling a suitable seed crystal from the feed material being melted at a temperature of about 1500°C by the concentrated radiant energy. They have been growing a variety of rare earth orthoferrites such as REFeO3 where RE=Er or Sm. They have also been studying the iron borates Fe_3BO_6 with cobalt and titanium substituents for some of the iron. As one alters the dopant concentration in this material, either a spin reorientation transition or a change from antiferromagnetic to weakly ferromagnetic behavior occurs on increasing the temperature.

Dr. Tsushima's major interest has been spin reorientation systems such as neodymium cobalt-five, NdCo5 and the rare earth orthoferrites. Such systems change from one magnetically ordered state to another magnetically ordered state when thermodynamical parameters such as temperature, magnetic field or pressure are changed. As one increases temperature, the hexagonal NdCo5 system changes from a basal plane anisotropy to an axial anisotropy while the REFeO3 materials show the reorientation of the spontaneous moments between the a-axis and the c-axis of a distorted perovskite structure. He has studied these transitions by means of Faraday rotation, magnetization, and specific heat.

Tsushima has found some interesting applications for these materials, it turns out many of the RECo5-type compounds exhibit such spin reorientation phase changes over a well defined temperature interval, $\Delta T = (T_{sr2} - T_{sr1})$. A large discontinuity in specific heat usually occurs at the upper limit, T_{sr2} of the temperature interval over which spin reorientation occurs. In particular, the $Dy_{1-x}Nd_xCo_5$ ternary system has such a transition whose

characteristic temperatures $T_{\mbox{sr2}}$ vary with its Nd concentration from 285 for NdCo5 to 367K for DyCo5. Tsushima makes use of the transition region, an interval of about 40K, where the material becomes magnetically soft, whereas above or below this interval the material is hard. By heating with a high intensity lamp or concentrated sunlight, a rod of this material, which is placed in a magnetic circuit containing a RECo permanent magnet bias and a transformer coil wound on one of the soft iron legs of the circuit, he obtains a device for converting thermal into electrical energy by suitably chopping the light. The $Dy_{1-x}Nd_xCo_5$ alloy is alternately changed from a soft to a hard magnetic state by the thermal impulses. The alloy thus modulates the magnetic flux linking the circuit as it changes from a high to a low permeability state. The device essentially makes use of the change in alloy specific heat that occurs with spin reorientation. This specific heat change represents about 10% of the total specific heat of the sample. It is about $10^7 \,\mathrm{ergs/cm^3}$. However, this total energy is not realized because only a surface heating is obtained at the chopping frequencies employed. Thus it is useful to use thin sheets of material having as large an area as possible. A chopping frequency of about 30 to 40 rpm is used. This effect has also been used in a photo-moto developed by Dr. Tsushima's brother at the Nihon Broadcasting Corporation, NHK. Elements consisting of orthoferrite sheets are fixed on the perimeter of a quartz wheel. The edge of this wheel is biased in the gradient produced by two sets of canted ferrite magnets located near the circumference of the wheel at either end of a diameter. The quartz disc is free to rotate on its axis and is caused to do so when chopped light is focused on the orthoferrite plates on its edge. Speeds up to 30 rpm have been achieved. Stacks of such disks on a given axis can be used to obtain a higher power output. This is a very novel device. A demonstration has been running in a department store window in Tokyo for several months with no difficulties. Rare earth cobalt biasing magnets could be used with Dy_{1-x}Nd_xCo₅ single crystal materials to obtain greater torque output from such a photo-motor. Some of this work has been written up in IEEE transactions on Magnetic MAG 13, 1158 (1977). A paper was also presented at the Hakone Workshop on this work. Dr. Tsushima showed us a short film demonstrating these devices.

Dr. T. Shibata and Mr. T. Katayama, with the help of Mr. M. Katsu of the Brother Industrial Co., have been studying the coercivity in $Sm(Co_{1-x}TM_x)_5$ sintered magnets where TM = Fe, Mn, Cr). A dramatic maximum in the coercivity, iH_C vs composition curves was observed at x = 0.1 in the Sm-rich compounds, $SmCo_{4.35-x}TM_x$ for each of the substituents. Sharply peaked iH_C vs annealing temperature curves were also found with the optimum temperature in the range 850-900 C. They felt that the coercivity mechanism in these materials was the same as that for $SmCo_5$, namely, reverse domain nucleation. The details for this work were given in Paper XI-5 at the Hakone Workshop.

The orthoferrites are also being studied for use as optical modulators. A thin single crystal is biased in a Helmhaltz coil set. When helium-neon red laser light impinges on the crystal after passing through a polarizer, it is transmitted through the analyzer when no magnetic field is applied. In a field the plane of polarization is rotated so that no light is transmitted through the analyzer. Fields of only 10 to 30 Oe are used to modulate the light. Good contrast is produced. To date only modulation frequencies up to 1 kHz have been used. But the high domain wall velocities indicate that the orthoferrites should be capable of being modulated to frequencies as high as 10 MHz. This is an order of magnitude better than can be achieved in the parnets.

20

We were given a tour of all the laboratory facilities. The most interesting work that I saw was on a time-resolved observation system for the high speed contracting motion of bubble domains in real time being done by Dr. M. Hirano. A TV system was used for recording the motion of bubble domains generated by the incidence onto LPE-garnet films of laser pulses produced by a Q-switched YAG:Nd³⁺ laser that puts out 10 µsec pulses. These cause domain stripes to move across the field of view. One can get the velocities of these stripes from the pictures that they take. They had to modify the electronics of the TV system so that instead of getting 16 or 32 frames per second they choose one particular frame to record magnetically each second. They have found that the contracting velocities in YIG-coated films have an assymmetry with respect to the sense of the in-plane field. This assymmetry can be attributed to the YIG layer being on one side of the films. The details of some of these experiments were published in J. Appl. Phys 49, 1909 (1978).

The Magnetics Section is also doing work on magnetic semiconductors. Large (~ 1 cm diameter) single crystals of CdCr₂Se₄ have been grown using a traveling-zone, flux-melt technique. These are huge compared to the best size previously available, which were cubes about 2 mm on a side. There is no practical use yet foreseen for this work. Various magneto-optical and resonant Raman scattering studies are being done to elucidate the electronic structure of this spinel-type ferronagnetic semiconductor. At the moment, it is just a basic research study.

Silicon substrates are being used to grow their Gd-Co-Mo amorphous films for bubble memory work. Of course, the single crystal Si substrates are easy to obtain; they have a good thermal match to the GdCoMo material and are much cheaper than the GGG, gadolinium gallium garnet commonly employed as a substrate.

It was a very interesting visit and Dr. Tsushima was a very gracious He also arranged for me to speak to some of the people involved with the energy applications of hydrogen. As mentioned above, ETL has a major mission to develop the solar and hydrogen energy technologies under the national "Sunshine Project." I received a brochure in English describing this Project from the Chief of the Energy Systems Section, Dr. T. Horigome. He also gave me a paper describing the general aspects of R&D on solar thermal power systems in Japan. It described the two different types of pilot plant now being studied, namely, the Tower type or Central Receiver type and the Plane-Parabola type. The former is suited for a large capacity plant and the latter for a relatively small scale application. I was also introduced to Dr. S. Ihara who is the project leader of a project to produce hydrogen from water by using solar thermal energy. He gave me two papers describing his work. One was a reprint entitled "Feasibility of Hydrogen Production by Direct Water Splitting at High Temperature," Int. J. of Hydrogen Energy 3 287 (1978) and the other was a preprint of a paper "On the Study of Hydrogen Production from Water Using Solar Thermal Energy," given at the 2nd World Hydrogen Energy Conference held at Zurich in 1978. His analysis of overall efficiency and target cost shows that the process would be justified for practical use if an effective method for separating the H2 and O2 gases at high temperatures (\sim 3000K) can be developed. He points out that the barrier is the lack of a suitable non-porous membrane for selective separation of H2 from O2. Such a membrane has not yet been

demonstrated in the high temperature range where substantial decomposition takes place. That has tested materials such as Nb, Ta, W, $\rm Zr_2O_3$ - CaO, and $\rm Al_2O_3$ by using a Xenon-arc high temperature image furnace to simulate a solar collector. A small amount of hydrogen has been separated from dissociated water vapor at about 1800K. However, the feasibility of using these materials as membranes is still uncertain, since the metals are chemically unstable in water vapor above 1000K, and the ceramics are easily broken by thermal stress.

30 May AM - Visit to the Toshiba R&D Center, Kawasaki

Personnel Contacted:

Dr. Shu Chiba, Manager Metals and Ceramics Laboratory

Mr. Naoyuki Sori, Deputy Manager, Metal Engineering Section

Dr. Koichiro Inomata, Senior Researcher

Mr. H. Hoxie, Researcher

Mr. Teruo Oshima, Researcher

We were greeted by the above people in a rather handsome conference After we were served tea, Dr. Chiba showed us a brief closed circuit TV film of a few of the projects being worked on at the Toshiba R&D Center. This work included e-beam lithography using both positive and negative resists with resolutions better than 1µm; lithium tantalate SAW devices; the technology of fabricating NbTi superconducting wire; an in situ method for producing Nb3Sn superconducting films; projects concerning VLSI; and superconducting levitated high speed transportation systems. Toshiba, Hitachi, Mitsubishi, Matsushita are all collaborating with Japan National Railway to produce a superconducting levitated train. Such a joint effort would be impossible in the USA because of our anti-trust laws. Synchronous pulsed coils in the track supply the energy for levitation and propulsion. Superconducting magnets are located on the train cars. The train becomes levitated at speeds above 100 km/hr. Current designs call for a maximum speed of 500 km/hr. We were told that Toshiba has about 60,000 employees of whom 1,500 work at the R&D Center.

After the overview by Dr. Chiba, we then met with Dr. Inomata and his associates to discuss the Toshiba REPM effort which primarily addressed the development of company products. These include high-fidelity loudspeakers and earphones, stepping motors for clocks, radially anisotropic "TORSOREX_{TM}" magnets for motors and generators, magnetrons and large generators. They showed us new, very thin loud speaker designs that were now in production. They were quite proud of a new REPM-based magnetron tube for their microwave cookers. This tube replaces one that used a ferrite biasing magnet. The new tube has an output of 1 kW as compared to the 0.75 kW of the old design. It also has about two thirds the weight and volume of the old tube, which had occupied a cubic volume 4" on a side. All of their magnets employ Ti rather than Zr as the substituent used to enhance the coercivity. They are making cost-effective cerium-based magnets with an energy product close to 14 MGOe. They showed us anisotropy constant data on the system $Ce(Co_{0.851-x}Cu_{0.136}Fe_{x}^{Ti}_{0.013})_{6.4}$. A maximum occurs at X 0.13 with a value of K=2.5x10⁷erg/cm³. We were given a large, as-cast sample having a (BH_{max}) ± 13.6 MGOe and a composition $Ce(Co_{0.742}Cu_{0.14}Fe_{0.107}Ti_{0.011})_{6.32}$. Their samples are induction melted under an argon atmosphere in magnesia crucibles and cast into an iron mold.

The as-cast ingot is then ground into powder without a homogenizing heat treatment. The powder is subsequently aligned with a pulsed magnetic field, pressed into the desired shape and sintered at 1100°C in an Argon atmosphere for an hour. It is then given a controlled air quench with rapid cooling rates of about 200 to 300° C/sec followed by a one hour anneal at 400° C to develop the intrinsic coercivity IH_C . IH_C remains at about 5.5 kQe independent of annealing temperatures in the range 300-700°C. Above 700°C it drops rapidly to below 1 kOe. They indicated that this material has a 1:7 composition in the grains and 1:5 in the grain boundaries. Dr. J. Livingston of G.E. has analyzed the microstructure of the Toshiba Ce-based material and published the results in IEEE Trans. Magnetics, MAG-10, 313 (1974). Their higher energy product samarium cobalt-based alloys are close to the 1:7 composition and also contain Ti as the coercivity enhancing substituent along with iron and copper. Dr. Inomata also gave me samples of their TOSOREXTM, TS-22 Sm-Co material, which is rated for continuous use to 200°C. At 230°C the material shows a loss of only 5% in its remanence. Inomata was interested in having us measure the anisotropy fields in their materials.

Having overseen the development of comercially useful magnet materials. Dr. Inomata has now been assigned to the development of amorphous metal, high permeability magnetic materials. This is unfortunate from the point of view of fostering a basic understanding of the REPM materials, since he had been doing some very important NMR studies concerning the contribution of the various cobalt sites to the magnetization and anisotropy of these materials. As was mentioned above, Prof. Wallace feels that the most revealing theoretical treatment of the contributions to the anisotropy by the different Co sublattices has been made by Dr. Inomata, who has not yet felt confident enough to publish his results. Inomata gave me a sample of the new amorphous magnetic material that his group has developed to replace permalloys in recording heads. It has the composition -(Coo.88Fe0.06Ni0.03Nb0.03)75Si10B15. The Ni and Nb substituents are used to enhance the mechanical stability of the material. I promised to check on what effect the absorption of hydrogen might have on the magnetic properties of this material, since Dr. Tauber and I have found that other Metglass-type materials can have their magnetic properties significantly degraded or enhanced by the presence of hydrogen.

30 May PM - Visit to the National Institute of Metals, Tokyo

Personnel Contacted:

Prof. - Dr. Toru Araki, Director

Dr. H. Maeda

Dr. H. Moma

Dr. Y. Sasaki

Dr. K. Hoshimoto

Prof. T. Araki, the Institute Director, and Dr. Maeda, our host, met with us in a nicely furnished conference room over the customary cups of green tea to give us a history of the Institute and an overview of the current projects. Prof. Araki has had a distinguished career dealing with the metallurgy of steel. He is currently President of the Iron and Steel Institute of Japan. The Institute of Metals publishes its own journal in English, "Transactions of the National Institute of Metals." A subscription

can be obtained by writing to Prof. Araki. The Institute is also participating in the national "Sunshine" and "Moonlight" projects. The first is an effort to develop new energy sources, while the second is to extend and promote energy conservation methods. There is a major effort on the development of various corrosion and stress resistant steel alloys for the shipping industry. The improvement of welding techniques for steels and the tensile testing of welds are also areas given great emphasis. Other areas of interest are the rare earth cobalt permanent magnets, metal hydrides for hydrogen storage applications, and amorphous metals. We were told that the Institute will be moving within a year or two to modern facilities in the new "Science City" Tsukuba about 120 km north of Tokyo. A new university is to be located there as well as many of the national laboratories.

Dr. Maeda took us on a tour of the Institute facilities which were housed in a large complex of buildings about 60 years old. Everything seemed to be well-maintained. We were shown some huge tensile testing machines about two stories high used for testing the single pass welds in two or three inch thick steel plate used for building the supertankers. Also quite impressive was a floor filled with row on row of high temperature stress corrosion furnaces used for the evaluation of steels and other high temperature alloys to be used in the eventual construction of breeder reactors. These computer controlled and monitored furnaces were part of a five year, fifteen million dollar testing program to determine the at-temperature, stress corrosion properties of many alloys. They feel that only data taken under actualuse conditions for long periods of time are valid. They don't believe that the accelerated testing and statistical modeling used by the Americans have been valid approaches as evidenced by the large amount of down-time in our reactors due to the failure of steam lines. There were eleven hundred such furnaces in use on one floor and seven hundred and fifty on another as part of this extensive testing program. These tests are under the direction of Dr. Moma and involve following the interaction between creep and fatigue, especially thermal fatigue, and employ the alternate low-frequency application of compressive and tensile stress at high temperatures.

Dr. Maeda told us briefly about the coercivity calculations that he has done for the highly anisotropic $R(Co_{1-x}M_x)_5$ alloys where R=Y, Ce, Pr, Sm and M=Cu, Ni. He used a simple discrete-spin configuration model for a domain wall in the presence of planar short-range fluctuations of the exchange interaction and anisotropy, which strongly depends on fluctuations in the distribution of non-magnetic elements. He has found that by using a reasonable magnetude for short-range fluctuations he can predict coercive fields of the same order as those measured.

Dr. Maeda introduced us to Dr. Y. Saski who has developed a fine facility for studying the hydriding of magnesium-based alloys for use in automotive applications. He has a Kahn balance arrangement, such as Dr. A Tauber is building at ET&DL, for studying hydriding kinetics at various temperatures for pressures below one atmosphere. The pressure below one atmosphere is regulated by the thermal decomposition of ZrH2 in a temperature-controlled reservoir. He also has a high pressure apparatus for doing similar studies in pressures to 70 atmospheres. This unique apparatus was built under contract by Japanese industry and employs a hydrogen cylinder with a mechanical compressor. Sasaki has been studying hydriding in the single phase Mg2Ni alloy and the polyphase material Mg0.60Ti0.15Ni0.20Fe0.05. Some

of Dr. Sasaki's work on these metal hydrides is being sponsored by the Daimler-Benz Company of West Germany in connection with their attempt to develop a hydrogen-fueled automobile.

We were also introduced to Dr. K. Hoshimoto who has been studying the hardness and tensile strength of amorphous metals of the system $(\text{Fe}_{x}\text{Ni}_{1-x})_{78}\text{Si}_{10}\text{B}_{12}$. His work correlating the microstructure of these materials as seen in electron micrographs with their hardness and tensile strength was scheduled for publication in Scripta Metallurgica. He was nice enough to give me samples of his alloys so that we might check on their capacity to absorb hydrogen and any consequent changes in their magnetic properties.

31 May - Visit with Dr. Justin Bloom, Scientific Counsellor, U.S. Embassy, Tokyo

At the request of Major O'Brien, I visited with Dr. Bloom to give him an overview of the Permanent Magnet Workshop held at Hakone. After giving him a summary of some of the major technical results, I commented on the concern expressed by many of the American attendees about the commanding lead that the Japanese have taken in this field. He indicated that he had already received calls from a few representatives of American Companies expressing their distress about the situation. He also said that a similar problem exists in nearly every aspect of technology and that he had been sending messages of concern to his superiors at the Department of State for several years, but to no avail. He asked if I would make my observations known to Ambassador Thomas Pickering, Assistant Secretary of State for Oceans, Environment and Science or to Mr. Alan Romberg, Director of the Japan Desk, if they requested a briefing. Bloom felt that the impressions of a practicing scientist might have more influence than his own reports have had. I subsequently received a copy of a teletype that Bloom sent to Pickering suggesting that I be contacted. However, I never heard from the State Department. Bloom was interested in receiving a copy of my trip report when it became available.

1-2 June - Visit to Shin-Etsu Chemical Industry Co., Takefu, Fukui

Personnel Contacted:

Dr. Y. Tawara

Mr. M. Honshima

Mr. T. Chino

Mr. K. Ohashi

Mr. C. Nakazawa, Director

On Thursday evening we were graciously met at the train station by Dr. Y. Tawara and his associates who took us to our hotel. We then went out to a pleasant dinner with Dr. Tawara and his family. The next morning we went to the Shin-Etsu plant where we were greeted over tea by the Director, Mr. C. Nakazawa, who told us about the Takefu facilities and their products.

The Shin-Etsu plant at Takefu employs about 600 people. They are engaged in rare earth oxide reduction, rare earth separation and the production of rare earth-cobalt magnets. They also produce polypropylene thin films for capacitors and single crystal silicon for the semiconductor industry. Their subsidiary Shin-Etsu Hannotai was formerly a partner with Dow Chemical and

today is one of the world's big three producers of single crystal silicon (along with Wacker and Monsanto). Shin-Etsu also produces fused quartz for semiconductor processing furnaces, and high quality quartz for optical fibers. The quartz products are made at a plant in Esobe. Silicone oils and rubber are also produced at the Takefu location. After giving us an overview of the Shin-Etsu business, Mr. Nakagawa left us to detailed technical discussions with Dr. Tawara and his rare earth magnet group. These talks were very open. They furnished us with many technical details which they have not yet published in the open literature. Even the subsequent trip through their manufacturing facility was quite open. They answered all technical questions most freely. This frankness was due to the friendship between Prof. Strnat and Dr. Tawara, who had spent nearly a year as a visiting scientist in Strnat's laboratory at the University of Dayton a few years ago.

The raw material for their magnet and rare earth production is obtained from Europe as a concentrate from xenotime ores. They obtain Y. Sm. Gd and a small amount of Eu. They use an ionic separation and solvent extraction method to obtain the rare earth metals. Since their own production of samarium is insufficient for their magnet production needs, they buy Sm metal and samarium oxide where they can in the world markets. They make their magnet alloys both by a direct alloying of the metals and through a reduction-diffusion processing of the oxides. They are presently making both Ce-based and Sm-based magnets. They are using about 15 tons/year each of Sm and Ce. The cheaper Ce magnets are used in watches and clocks. In fact, the Japanese are exporting watch movements to Europe, even to Switzerland and France. There is a much bigger demand for the Sm- than for the Ce-based magnets. However, since Rhone-Poulenc, a major rare-earth supplier, has warned about the limited world supply of Sm, the Shin-Etsu people are trying to conserve Sm by combining it with Ce while still trying to maintain a relatively high energy product magnet. Thus, while the compound CeCo5 has only a (BH)max of 15 MGOe, the compound Sm0.5Ce0.5Co5 has (BH)max=20 MGOe. With proprietary substituents into this latter material the Shin-Etsu people expected to be in production with a 22-MGOe magnet in about three months.

They are also developing Sm_2Co_17 -type compounds having energy products close to 30 MGOe. Since the TDK patents for such materials are based on materials having $7.2 \le Z \le 8.5$ in the formula $Sm(Co,Fe,Cu,Zr)_Z$, the Shin-Etsu group is developing material with Z=6.2. They are also using Zr substituents to develop the coercivity. However, by using the lower transition metal concentrations they avoid conflict with the TDK patent position.

Dr. Tawara and his people still are uncertain as to how the Zr is enhancing the coercivities in these materials. I told them about my discussions with Takehashi and about my speculation that internal magnetic fields may be imposing order on a fine dispersion of ZrCo precipitates. They had no comments on this idea. We also discussed the idea of a ZrC precipitate contributing to the coercivity as put forth at the Hakone Workshop by Drs. Greinacher and Velicescu of the Goldschmidt Co. Dr. Tawara said that it is difficult to keep the carbon level down in pure Sm or Sm2O3, since some contamination is inevitable from the milling with hexane or toluene. Greinacher of Goldschmidt claims there is always some carbon contamination in the starting oxide materials. Tawara said that while CO2 goes off during the arc melting process so that only 0.002% carbon is left.

this concentration goes up about tenfold after milling since milling brings in CO₂ from the air. Thus, the Shin-Etsu people specify a 0.02% carbon concentration in their powder materials. Some further addition may take place in the pressed magnet bodies due to the stearate-type mold release employed which enters the magnet during the sintering process.

Dr. Tawara pointed out that Shin-Etsu has been using Mn to enhance the coercivity of their magnets for some time, even though they have never published or said anything publicly before. They have studied the system Sm(Coo.80Fe0.05-vCuo.15Mnv)6.85 and found they got fairly good magnets with a (BH) max of 23 MGOe with y=0.03. The intrinsic coercivity THC is found to increase fairly uniformly with Mn concentration even up to y=0.05, i.e., with no Fe present. While a quench gives higher THC, they usually age their material at 750°C for about 45 minutes and then give it a slow furnace cool to 400°C with a final cool, also in an argon atmosphere, to room temperature. The slow cooling gives sound magnets with uniform properties. One of their present commercial magnets has a energy product of 21 MGOe with y=0.025 and is made with a slow cool down. Another magnet compound, their S-25 material, has a $(BH)_{max}$ =25 MGOe with Z=6.8. It contains Ti as well as Mn and has the formula $Sm(Co_{0.702}Fe_{0.15}Cu_{0.11}Mn_{0.03}Ti_{0.008})_{6.8}$. The Ti is used as a replacement for Zr. The combination of the Mn with the Ti helps Shin-Etsu get around the Toshiba and TDK patents.

We were taken for a tour of the magnet measurement and production facilities. Their measurement facility includes an electromagnet capable of achieving about 18 kOe. They have an automated B-H Analyzer with an analog-to-digital tape data storage system which used an Intel 8080 16 kb RAM. The system, designed and built by the Shin-Etsu Engineering Co., uses two tape cassettes - one for data storage and the other for programming. The system is used as a permeameter with the magnet poles screwed down flush to the magnet face so that demagnetization effects are eliminated. A search coil surrounds the magnet and its output is fed to the integrating flux meter assembly.

A pulsed-field magnetizer (2,500 V, 2,000 μF) capable of supplying a 15,000 A pulse with a l ms rise time and a broad l ms peak was used for the one shot magnetization of a variety of magnet configurations. Several different coil jigs were available to supply axial, radial or transverse magnetizations. The peak pulsed magnet field achieved depended upon the jig coil configuration but usually was about 25 kOe. They also had a 50 Hz demagnetization arrangement for the 2-17 type materials because of their relatively low coercivities. All production magnets are pulse magnetized, tested and then demagnetized before shipment to the customers.

Sintering and heat treatment of magnets is done in three large, 12 ft long furnaces which are heated with carbon resistance elements. An argon atmosphere is used in all the operations. Each furnace has three chambers held at the different temperatures for a batch processing, semicontinuous operation. They also have several small test furnaces, 8 ft long, also with three chambers separately controlled. Green-pressed magnet material is first preheated at 400°C and then sintered at 1200°C. After sintering, samples are moved through gate valves with fork-lift type transfer devices to a fan-cooled chamber. The S-25 material is given a subsequent treatment in an aging furnace 8 ft long which has four chambers at temperatures of 750°C, 650°C, 550°C and 400°C. This furnace has tantalum windings and also uses an argon atmosphere. A long push-rod is used through a vacuum seal to push the molybdenum sample boats, having inconel edges to prevent warping, to the various heat treatment zones.

Automated multiple punch-presses with pulse electromagnets associated with each die were used to align and press the magnet powders. Various dies were available to make bars, rings and radial arc segments. Two types of presses were available, either with the aligning field perpendicular to the pressing direction or parallel to it. The aligning fields used were about 10 kOe. For axial (parallel) pressing the pressure used was about 1 ton/cm² while for transverse pressing it was 0.6 ton/cm². The latter was the method for making their large standard blocks (1x3x6 cm). The green density achieved was 54% of the theoretical value while the final density after sintering was 99.8%. The die lubricant was stearic ac.

The factory is working 24 hours per day, seven days a week. Four full-time production teams have been trained and they rotate their shifts from week to week. Shin-Etsu is currently selling all the magnets and magnet powder they can make.

Both attritor and jet milling processes are used to make the magnet feed powders. The jet milling is more convenient for the Sm-based magnets, while the attritor is more useful for the Ce-based material. At the moment they have one large unit of each type and are in the process of acquiring another jet mill. These units start with coarse 20 mesh powder obtained from another plant in Takefu. The jet mill can process material at the rate of 150 kg/day to produce the final 3 µm powder. The attritors and jet mills are made by Japanese companies. The jet mill was made by the Japan Pneumatic Co., while the attritor was supplied by the Mitsui Mill and Attritor Co. The powder is sized by a Fischer unit. Six storage chambers are used for drying the powder by means of aspirator pumps. The powdered material is stored in sealed chambers under dry nitrogen gas. Final powders of various materials are stored in color coded cans in a fireproof cinder block room.

The final mechanical finishing of the magnets was done in a large modern automated machine shop. They had twelve automated cutting machines with sets of parallel diamond-impregnated blades of thickness from 0.6 -0.8 mm. These machines were used to square-up the ends of bar magnets. The cutting edges were on the inside surface of the blades. Automated surface grinders with magnetic chucks were used to attain given thickness specifications. Inside/outside grinders for segmented-arc, concentric torque coupler assemblies for large seal-less chemical pumps were also in use. Tight eccentricity tolerances are required for the torque couplers. Shin-Etsu is manufacturing torque couplers having two sets of eight magnets facing each other on concentric rings. The I.D. of the outer ring is 95 mm while the O.D. of the inner ring is 75 mm. The magnet dimensions are 5.5x5.5x1.0 cm and are made of the Ce-based Rarenet H material. The device delivers a torque of 180 kg-cm. A similar coupler employing the Sm-based Rarenet S-22 material delivers a torque of 300 kg-cm. Dr. Tawara indicated that larger torque couplers with a capacity of 3,000 kg-cm are being developed in Germany for autoclave chemical processing. The machine shop works only two shifts for 16 hours a day and handles about one-half of the machining load. The rest is contracted with outside Japanese companies. Incidentally, great care is also taken to salvage and recycle all of the scrap metal resulting from the magnet machining operations.

At this time Shin-Etsu is using about 50% of its magnet production/sintering capacity. They are now producing 3 tons per month which is about 50% Ce-based and 50% Sm-based. While the magnet business was profitable last year, it is not currently profitable because of the dramatic rise in

cobalt prices (nearly sixfold from \$6.85 to \$40 per pound) that has taken place since the invasion of Zaire in May 1978. Also the competition (TDK, Toshiba, Hitachi, Sumitomo, Mitsubishi) is keeping magnet prices down. Market prices are still going down in Japan. Shin-Etsu is carefully thinking about recycling processes to salvage solid material from breakage in handling the brittle magnets, machining, and cutting sludges from water grinding operations. They are also seriously considering producing plastic bonded magnets which would not be so brittle and could be pressed to any size and shape.

4 June AM - Visit to Sumitomo Special Metals Co., Ltd., Yamazaki Works, Osaka

Personnel Contacted:

Dr. A. Higuchi

Mr. Y. Matsuura

Mr. N. Ishigaki,

Mr. H. Yamamoto

Mr. Aoyagi, Director, Manager of Yamazaki Works

Dr. Higuchi was kind enough to meet us at the Kyoto railroad station on Sunday afternoon and take us to our nearby hotel. He later joined us that evening for a pleasant dinner with his wife and her lady friend who spoke English. The next morning his wife and her friend took Mrs. Strnat shopping while we accompanied Dr. Higuchi to the Yamazaki Works in Osaka for technical discussions and a tour of their plant. We were received by the Director, Mr. T. Aoyagi who told us about the history of the Sumitomo Special Metals Co. and the products it makes. They were making ferrite magnets at a rate in excess of 1,000 tons/month. They are second only to TDK and are aiming to achieve a production of 1,500 tons/month at their Kyushu plant. They presently produce about one third of the Japanese ferrite magnet output. A new fully automated plant was also due to go into production in July 1979 at YABU in Hyogo Prefecture. They hope to achieve with this plant on-line an ultimate total production of 3,000 to 4,000 tons/month. Sumitomo also makes cast and sintered nickel based alloys, permalloys, glass-sealing Kovars, FeCrCo permanent magnet alloys, and 45% of the Japanese Alnico production. Their Alnico production rate was currently 200 tons/month with a capacity for 500 tons/ They are also producing the RE cobalt permanent magnets as well as such soft alloys as the permandurs and amorphous metals. They are using hot isostatic pressing methods for making soft ferrite materials for recording heads needed in computer, audio and video applications. Sumitomo is not involved in making any microwave or millimeter wave devices as such.

Their nickel alloys are produced at the Suita Works which has facilities for hot and cold rolling. Layers containing three to five sheets can be bonded simultaneously by cold welding in one pass. Such composites replace pure Ni in various anode applications, especially in power transmitting tubes for the television industry. The same facility produces Nichrome and Alumel as well as the low expansion coefficient Invar and Superinvar alloys.

The Yamazaki Works makes a variety of Alnico and RE cobalt magnets for torque couplers, TWT's, magnetic separators, and toner arms for Xerox type photo-copiers. They also make a sheet floating device using permanent magnets for handling large steel sheets in various manufacturing plants. They also use micro-computers such as the Hewlett-Packard 9820A to design magnetic

circuits for their customers. In fact, an automated hysteresograph, which uses an HP9820A, was designed by Dr. Higuchi and is being marketed by the YHP Co. (Yokagawa-Hewlett-Packard) in Japan.

In the manufacture of the RECo magnets, facilities for x-ray fluorescence and wet chemical analysis are used for quality control. In particular, the hydrogen, oxygen, carbon and sulfur content of the starting powders are monitored.

Mr. H. Yamamoto is working with (Mischmetal)Co5 magnets trying to optimize the substitution of Fe for Co and Sm for MM. He finds that the coercivity critically depends upon the milling and sintering procedures. SmFe5 and MMFe5 have not been observed,only the 2-7 and 2-17 phases seem to form. Al Ray commented that the systems RE Co5-xFex form only for X = 0, when RE = Sm and for X $\stackrel{\leq}{=}$ 1.5-2 for RE = Y or Ce. Namiki in Tokyo claims to have made a reasonable (Sm1-xYx)2Co17 magnet where X = 0.1. They have licensed Sumitomo to make such magnets.

After a brief luncheon with Dr. Higuchi and Mr. H. Yamamoto we left to visit the Matsushita Electric Industrial Co.

4 June PM - Visit to Matsushita Electric Industrial Co., Ltd. - Materials Research Laboratory - Moriguchi, Osaka

Personnel Contacted:

Mr. Y. Sakamoto

Dr. T. Kubo, Head of the Magnetics Division

We were graciously received by Mr. Sakamoto and Dr. Kubo in a small conference room where many magnetic devices employing manganese aluminum carbon, MnA&C, magnets were displayed. Matsushita has developed this unique magnet material that has an energy product of 6.0 MGOe and a coercivity of about 2.8 kOe. This standard alloy, now being sold commercially, has a nominal composition of 70 W/o Mn,29.5 W/o Al, 0.5 W/o C. However, an energy product of 8.0 MGOe has been achieved in the laboratory with a small amount of Ni substituent. Sakamoto indicated that the material had a relatively low Curie temperature near 300°C. The irreversible loss in magnetization is about 5-7% on cycling the material from room temperature to 100°C and back for an operating point at $(BH)_{max}$. No policy on licensing agreements had been established as of the time of our visit. Matsushita is the only producer of the material. They have a fully automated pilot plant on-line which produces several hundred extruded cylinders per month. A hot extrusion process at 700°C is used to achieve an 85% reduction in cross-section and to impart a uniaxial anisotropy to the material. Starting 80 mmd billets are cast in magnesia crucibles in air at about 1450°C. Very little oxidation occurs, but about 1% hydrogen is detected, since the starting electrolytic Mn contains hydrogen. Long bars about 31 mm in diameter are produced since Matsushita is using discs 31 mm d x 6 mm as speaker magnets. Several thousand test speakers have been produced for a new line of very thin (010 cm) Panasonic wallet sized AM-FM receivers.

The extruded MnALC material is very hard to machine and requires tungsten carbide tools as well as a special heat treatment. The material cannot be soldered to directly with lead-tin solder. But the use of an electrolytic coating permits such soldering.

The pricing of the material is between that of the Alnicos 5 or 8 and the ferrites. It can replace Alnico 8 for small magnet applications. This is important in view of the high cobalt costs which have driven up the prices of Alnicos.

The Matsushita people showed us several prototypes of new products which will use these magnets. These included:

- (1) a magnetic reed-switch security system.
- (2) a reed-tuning fork assembly for clocks.
- (3) an automotive engine speed tachometer.
- (4) an 8-pole magnetic toner-roller magnetized along four equally spaced diameters for xerox-type photocopiers. These 14 mm d rollers replace 41 mm d ferrite rollers. Here 2 MGOe isotropic magnets are used. These are produced by control of the composition, conditions of extrusion and heat treatment.
- (5) a flat, 6-pole, disc-armature motor for an electric bicycle. The MnALC replaces a ferrite used in a previous prototype and yields a 50% reduction in total motor weight and produces 1.5 times the torque. The bicycle has a 20km range and uses a lead-acid battery which can be recharged overnight. It is currently being test-marketed in the Osaka area.
- (6) a small 3" speaker for the "Mister Thin" Panasonic radios mentioned above. For a 3" speaker with comparable performance the use of MnALC requires a magnet weighing only 23 grams, while Alnico 5 and ferrite magnets would weigh 34 g and 48 g, respectively.
- (7) Various electrical analog instrument meters.

Dr. Sakamoto promised to send me samples of this material so that we could study the temperature dependence of its coercivity and anisotropy, especially in the interval between 4.2K and 300K where little data exists.

After our technical discussions Mr. Sakomoto took us on a tour of the Matsushita Electric Science and Engineering Exhibition Hall, a two-story building devoted to exposing the public to the advanced technologies being developed by the company. The many exhibits of new prototype products were most impressive. These included the following:

- (1) Portable Home Video Tape Recorder with a 6-hour playing time.
- (2) "Pana-Puel_{TM}" Catalyst for Exhaust Gas Purification uses inexpensive compounds of iron, manganese and zinc solidified with a calcium aluminate cement and is suitable for home cooking and heating appliances as well as for industrial pollution control.
- (3) Honeycomb Disc Diaphragm Speaker Systems Uses a flat diaphragm of honeycomb boards containing thousands of hexagonal cells to replace conventional paper cones which give undesirable resonances. This allows reproduction of sound over a wider frequency range than conventional systems by approximately 2 octaves along with a flat frequency response.

- (4) "ANOGROM_{TM}", New Metal Recording Tape Matsushita has developed an evaporated metal recording tape and is marketing it as a 3-hour microcassette tape.
- (5) High-Speed International Facsimile System UF-SOSG. This unit is the world's first facsimile unit to meet the International Standard (Gll1) for high-speed facsimile transmission. It can transmit a legal B-4 document with approximately 700 characters in only 20 seconds to any Gll1 standard unit in the world.

In addition, they had an impressive array of solid state devices on display such as:

- (1) TV camera tubes using Z_nSeTe detectors.
- (2) Low light level TV cameras with ZnCdTe detectors.
- (3) LiNb0, and NH_2PO_A piezoelectric transducers.
- (4) Lithium batteries for fishing buoy lighting systems.
- (5) Electrolytic capacitors with the remarkable rating of 10 farads at 1.6 volts d.c. These were only 3/4" 0.D. and 2" long!
- (6) Sensors of various kinds for gases (natural gas, Cl₂, SO₂); Hall detector IC chips; SiC thermistors.
- (7) SiC surge arrestors.
- (8) Flat, disc-armature AC-DC motors.
- (9) FeNi high energy density batteries.
- (10) Paper thin, low-drain batteries made of stainless steel and a perchlorate compound.
- (11) Closed circuit TV-I.D. card production system (now being used by the New York City Police Department).
- (12) MnAlC magnets.

- (13) Computer-controlled home protection and regulation systems.
- (14) Computer based music synthesizer system.
- (15) Computerized learning centers.

The visit to the Matsushita facility was very stimulating. It was a fitting finish to a trip that left me overawed with the Japanese ability to take advantage of new research (much of which was done originally in the USA) and develop new, high-quality consumer products having mass markets.

SIGNIFICANT ACTIONS TAKEN

A. As a result of my concern that little or no work was being done by American magnet materials manufacturers to develop the new, higher energy product Sm₂Co₁₇- based materials that will be critically needed in new DOD systems, I arranged to address a meeting of the Magnet Materials Producers Association (MMPA) in Chicago on October 30, 1979. There, I met the executive officers of many of the companies involved with making permanent magnets. I was able to confirm that very little work is currently being done in the USA on the 2-17 materials. All companies expressed an interest in participating in an Army sponsored Manufacturing Methods Technology program to develop manufacturing capability for the 2-17 magnets, if such a program were initiated.

The Association voted to provide me with backup information on the current manufacturing capabilities for hard magnet materials. A question-naire was prepared and sent to the MMPA Executive Secretary, Mr. Thomas Dolan, for circulation to the membership. Such a compilation will be used in preparing the P-16 Justifications for the initiation of suitable MMT programs.

At the MMPA Meeting, I also learned of a severe problem that has arisen in the testing of the rare earth cobalt-based magnets. Many of the producers do not have the measuring equipment to measure adequately the properties and to characterize fully the rare earth cobalt magnets they produce. In fact, there is a serious lack of adequate, commercially available measuring equipment for evaluating the various magnetic aspects of these materials. Only a few university or government laboratories equipped with super-conducting magnets and specially designed, unique magnetometers have the capability for making such measurements in the USA. Magnet manufacturers are using rather qualitative evaluation methods for screening their materials at this time. They have no adequate means for the quality-assurance testing of the magnets or magnet assemblies that would be critical for projected DOD needs. The need for a Materials Testing Technology (MMT) program was also clearly indicated.

B. Visits, conversations and correspondence with various magnet users and producers have been initiated to better assess the DOD market for the REPM as well as our current and projected capability for supplying that market. Contacts have been made with personnel in the following organizations in the USA.

Naval Research Laboratory
Naval Electronics System Command
Air Force Materials Laboratory, WPAFB
Raytheon Co.
Thomas & Skinner Inc.
Brown, Boveri, Recoma
Hitachi Magnetics Corp.
The Charles Stark Draper Laboratory
Inland Motors
The Singer Co./Kearfott Div.
Colt Industries, Crucible Inc. Research Center
Electron Energy Corp.

- C. Steps have been taken to initiate the following MMT programs:
 - (1) High Coercivity, High Energy Product Magnets a three-year program to start FY-82, Total Cost \$1.4M.
 - (2) Intrinsically Temperature Compensated Magnets a three-year program to start FY-83, Total Cost \$1.25M.
- D. Also, the need for an MMT program identified above has led me to draft a MTT project request entitled "Rare Earth Cobalt Magnet Testing Technology" to be submitted for consideration in the FY-82 budget. The estimated cost would be about \$3.5M over a three-year period.
- E. A contract was let through ARO with Profs. K. Strnat and A. Ray of the University of Dayton entitled "Preparation and Characterization of High Energy Permanent Magnet Materials of the Type $\mathrm{Sm}_2(\mathrm{Co},\mathrm{Fe},\mathrm{Cu},\mathrm{T})_{17}$ " STAS agreements were let with the same researchers for the "Synthesis and Fabrication of Intrinsically Temperature Compensated $\mathrm{Sm}_2\mathrm{Co}_{17}$ Based Permanent Magnets."
- F. An in-house evaluation of the new Japanese permanent magnet materials having little or no cobalt was initiated. In particular, the anisotropic MnAlC samples supplied by Matsushita are being studied for the temperature dependence of intrinsic coercivity in the easy and hard directions.
- G. A Laboratory Research Cooperative Program (LRCP) was begun during the Summer of 1979 with Prof. David I. Paul, and his student, Ernest Potenzianni, of Columbia University. The objective of the research was to study, by means of Mossbauer spectroscopy, the effects of various substituents such as Zr,Cr and Mn on the hyperfine fields at the various Cobalt sublattices in the TDK type Sm₂ (Co,Fe,Cu,Zr,T)₁₇ type compounds. After a propitious start the program has been continued during the academic year at Columbia under a Post-LRCP grant from ARO. They are continuing their work at ET&DL during the Summer of 1980, employing a zero field NMR technique to compliment the Mossbauer work.

RECOMMENDATIONS

Many new applications are anticipated for the rare earth cobalt magnets in DOD systems. In addition to microwave/millimeter wave tubes and filters, these include novel cost effective accelerometers gyroscopes; fin-actuators for mini-RPVs, cruise missiles, anti-aircraft/anti-missile missiles and various combat aircraft; lighter, cost-effective motors and generators for combat vehicles, aircraft and torpedoes; as well as quadrupole lenses for particle beam weapon systems.

The Army is the only Service sponsoring any 6.1 work to promote this technology and that effort is only marginal compared to what is needed in this country. I recommend that this effort be significantly expanded and that a concomitant 6.2 effort be initiated.

I also strongly recommend that the MMT and MTT programs discussed above be promoted at the highest level to see that they be instituted, so as to assure that we have these strategic materials in the quantities required for the production of the advanced systems now being developed.

Finally, I recommend that we take a hard look at the Japanese methods for the management of R&D to see if we can learn from their successful techniques for creative interpersonal interactions. (A similar recommendation is to be found in a 20-page article on the problems with productivity and creativity in the USA that appeared in the 30 June '80 issue of Business Week.)

ACKNOWLEDGMENT

I would like to express my deep appreciation to all those named herein for their courtesy and cooperation during my visit to Japan to attend the Workshop, and during subsequent visits to Japanese magnet manufacturers. Additionally, I would like to thank the Japanese people for this aweinspiring glimpse of their culture: a culture unique in its deep and abiding respect for people, nature and material things.

APPENDIX

Itinerary:	
18 May 79	Depart Nwk 18:45 TWA Flight #127/Arrive San Francisco 23:20
19 May	Depart SF 10:00 Northwest Orient Flight #NW9 Arrive Tokyo 16:00 - 20 May 1979
20 May	Stay at the Sanno Hotel
21 May	A.M Visit Major O'Brien, US Science & Technology Center, Far East Office/P.M Train to Hakone
21-25 May	Attend 4th Int'l Workshop on Rare Earth Cobalt Permanent Magnets at Hakone Prince Hotel, Hakone
25 May	P.M Train to Tokyo - Stay at Sanno Hotel while in Tokyo
26-27 May	Visit in Tokyo with Prof. Strnat - Dr. K. Kamino of the Mitsubishi Co., & Mr. T. Kurino & Dr. H. Kaneko of the Society for Nonconventional Technology - Social/Sightseeing
28 May	Visit TDK Electronics Co., Tokyo - Mr. M. Ishikawa, Mgr. Magnetics Div. (with Profs. K. Strnat and A. Ray)
29 May	Visit Electrotechnical Laboratory, Tokyo - Dr. T. Tsushima, Dr. T. Shibata (with Profs. K. Strnat and A. Ray)
30 May	A.M Visit Toshiba R&D Center, Kawasaki - Dr. K. Inomata (with Prof. K. Strnat and A. Ray)/P.M Visit National Inst. of Metals, Tokyo - Dr. H. Maeda (with Profs. K. Strnat and A. Ray)
31 May	A.M Visit Dr. J. Bloom, Scientific Counsellor, US Embassy, Tokyo/P.M Train to Takefu, Fukui
1-2 June	Visit Shin-Etsu Chemical Industry Co., Takefu, Fukui - Dr. Y. Tawara (with Prof. Strnat)
3 June	Train to Kyoto/Osaka
4 June	A.M Visit Sumitomo Special Metals Co., Ltd, Osaka - Visit Dr. A. Higuchi (with Prof. Strnat)/P.M Visit Matsushita Electric Industrial Co., Ltd, Osaka - Drs. Y. Sakamoto and T. Kubo (with Prof. Strnat)
6 June	Depart Tokyo for Yokota AFB - Flight delayed 20 hours
7 June	MAC Flight #TKP-N2K6 13:00/Arrive Travis AFB, CA 5:45/U-Drive to San Francisco/Depart San Francisco 14:15 United Airlines #4616
7 June	Depart Newark via Limousine 23:45
8 June	Arrive Toms River, NJ 03:40

